Mathieu beams as versatile light moulds for 3D micro particle assemblies

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Abstract: We present tailoring of three dimensional light fields which act as light moulds for elaborate particle micro structures of variable shapes. Stereo microscopy is used for visualization of the 3D particle assemblies. The powerful method is demonstrated for the class of propagation invariant beams, where we introduce the use of Mathieu beams as light moulds with non-rotationally-symmetric structure. They offer multifarious field distributions and facilitate the creation of versatile particle structures. This general technique may find its application in micro fluidics, chemistry, biology, and medicine, to create highly efficient mixing tools, for hierarchical supramolecular organization or in 3D tissue engineering.

OCIS codes: (220.4000) Microstructure fabrication, (140.7010) Laser trapping, (350.4855) Optical tweezers or optical manipulation, (070.3185) Invariant optical fields.

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1. Introduction

Any application of laser radiation has its specific demand on the beam shape revealed in certain intensity or phase distributions, polarization, momentum and propagation properties. Thus beam shaping has been a field of interest since the invention of the laser. Besides mechanical, electro-optic and acousto-optical modulation techniques, today especially holographic modulation plays an important role, because it enables almost arbitrary beam shapes in specific working planes. In applications as laser resonators, optical fibers or non-linear photonics, particular solutions of the Helmholtz equation attract interest, because they offer, in addition to different transversal field distributions, stable propagation properties. The class of propagation invariant beams, distinguished by its expanded Rayleigh length, offers several applications in optical lattices, microlithography, atom optics and optical tweezers [1].

Structuring material with light is an established procedure. Designed light fields cut and weld 3D artificial materials as photonic crystals and metamaterials by ultra short laser-based material structuring [2, 3], modulate one to three dimensional refractive index changes in nonlinear optical material [2, 3], and organize particles into artificial structures from μm-size to single atoms in optical trapping experiments [4, 5, 6]. Scanning techniques [7] already enable to sequentially build 3D particles arrangements in step by step processes. To pass on to parallel methods, it is necessary to create light moulds, whose form can be directly transferred to the material [8]. For a powerful method, light moulds have to meet two criteria: transversal diversity combined with self-healing properties for longitudinal invariance to enable extensive structures. Both criteria have been separately demonstrated [9, 10]. In the approach presented here, we realize a combination of both criteria using propagation invariant beams with self-healing properties having complex transverse field distributions. Among propagation invariant beams, the class of Mathieu beams are especially well-suited due to their variety of transverse, non-diffracting structure that allows a variety of 3D applications for moulding light fields to organize particles or matter. We demonstrate the use of Mathieu beams as tailored light moulds for the 3D arrangement of spherical and non-spherical micro particles.

2. Mathieu beams

Mathieu beams are solutions of the Helmholtz equation in elliptical coordinates (η,ξ), which are given by $\eta = \mathrm{Im}(\mathrm{arccosh}((x+iy)/f))$, $\xi = \mathrm{Re}(\mathrm{arccosh}((x+iy)/f))$ and the eccentricity parameter f. In elliptical coordinates the 2D Helmholtz equation separates into two equations,

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which can be reduced to one differential equation by the substitution $\eta = \imath\xi$, resulting in Mathieu's equation:

$$\frac{d^2\omega}{dz^2} + (a - 2q\cos(2z))\,\omega = 0 \tag{1}$$

Mathieu's equation always has one even and one odd solution which are described by two parameters: the ellipticity parameter $q = f^2k^2/4$ (where $k = 2\pi/\lambda$) and the order m.

The transverse field distribution of even $\mathrm{M}_m^{\mathrm{e}}$ and odd $\mathrm{M}_m^{\mathrm{o}}$ Mathieu beams are given by [11]

$$\mathrm{M}_m^{\mathrm{e}}(\eta,\xi,q) = \mathrm{C}_m\mathrm{Je}_m(\xi,q)\,\mathrm{ce}_m(\eta,q), \qquad m = 0,1,2,3... \tag{2}$$

$$\mathrm{M}_m^{\mathrm{o}}(\eta,\xi,q) = \mathrm{S}_m\mathrm{Jo}_m(\xi,q)\,\mathrm{se}_m(\eta,q), \qquad m = 1,2,3... \tag{3}$$

Their outer appearance (Fig. 1, 2 (a)) is affected by elliptic nodal lines (defined by zeros of

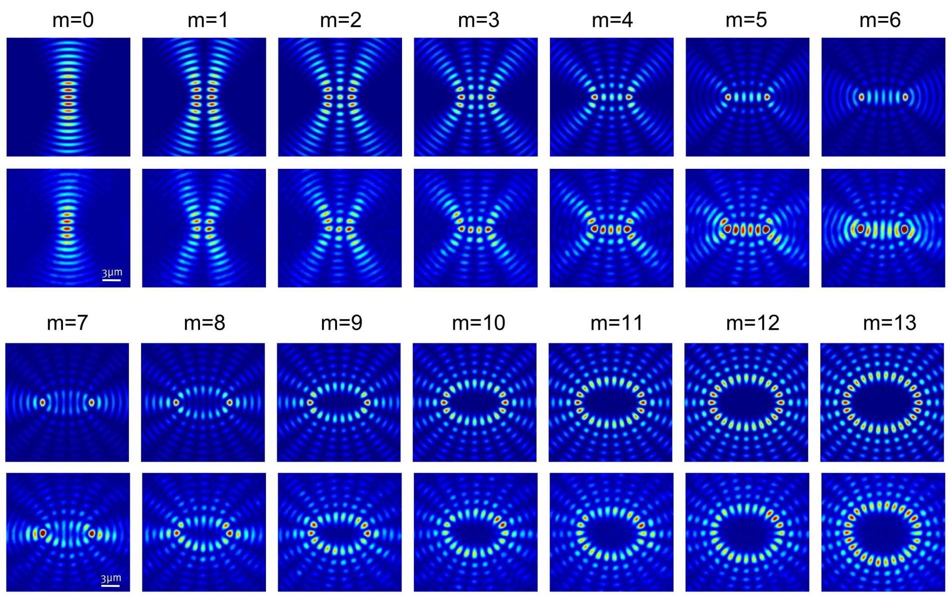

Fig. 1. Intensity distributions of even Mathieu beams (ellipticity parameter $q = 27$). Numerical calculations (upper rows) and experimental measurements with the optical setup described in section 3 (lower rows). All images are normalized to maximum intensity value and false colors are used to increase visibility.

the radial Mathieu functions) and by straight or hyperbolic nodal lines (defined by the angular Mathieu functions). Two coexisting solutions define even (Eq. 2) and odd (Eq. 3) Mathieu beams. The radial Mathieu functions Je_m and Jo_m are oscillatory functions, which have been described by Gutiérrez-Vega et al. [12]. The angular Mathieu functions ce_m and se_m are cosine-elliptic or sine-elliptic functions, respectively. C_m and S_m are normalizing constants. Besides even and odd Mathieu beams, superpositions of both as helical Mathieu beams [13, 14] (Fig. 2 (b)) are common.

Mathieu beams are self-healing beams, which rebuild their transversal intensity distribution if it is destroyed by any (small) obstacle. After a shadow zone, which depends on the obstacle's dimensions and the beam's angle with the optical axis, the beam is fully reconstructed. An estimate for the length of the particle's shadow zone $l = Rn/\mathrm{NA}$ is given by geometrical considerations [15]. The parameters are the particle's radius R, the refractive index of the

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medium n and the numerical aperture NA of the microscope objective.

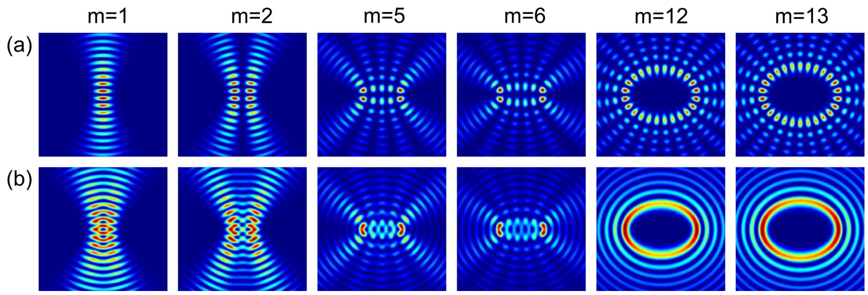

Fig. 2. Intensity distributions of a selection of odd (a) and helical (b) Mathieu beams (numerical calculations, ellipticity parameter $q = 27$).

3. Creating optical moulds with tailored light fields

We show the implementation of even Mathieu beams in optical tweezers and create 3D particle arrangements in the developed light moulds. To modulate amplitude and phase of the beam simultaneously, different techniques have been proposed [14, 16]. We multiply a blazed grating with the amplitude and add the phase to get the desired hologram. As is commonly done in holographic optical tweezers, we modulate the Mathieu beam by means of a spatial light modulator (SLM) in the Fourier plane of our optical tweezers setup. As for all propagation invariant beams, the field distribution in that plane lies on a narrow ring, effecting phase locked beam propagation, which is mandatory for this beam class. By implication, the diffraction efficiency in this configuration would normally be quite low. To overcome this bottleneck, we use a ring shaped hologram illumination as provided by an axicon [16], to maximise the available power. A second SLM can be used as an axicon, as we have done in our experiments (Fig. 3: axicon + lens), which makes us more flexible in adapting the hologram illumination. In this configuration, it is appropriate to unwrap the hologram, such that only the angular dependence is retained (Fig. 3: SLM 1). The radial dependence is then given by the pre-shaped illumination. This general technique to generate propagation invariant beams can be adapted to standard holographic optical tweezers setups, e.g. by replacing the final lens in a beam expander with an axicon. We additionally enhanced our setup with a stereoscopic microscope to directly take 3D images of the created structures [17].

The stereoscopic microscope illuminates the sample with two LEDs from different angles, generating two images, which are spatially separated into a stereo pair in the observation plane (Fig. 3). As an object moves axially in the sample, it appears to be displaced laterally in the two images, by the same distance but in opposite directions. The axial position can therefore be found by tracking an object in 2D in each image measuring the apparent difference in position.

After the linearly polarized laser beam is structured by SLM 1, the light is focused through an oil immersion microscope objective (M = 100x, NA = 1.3) into the sample chamber, where it generates the propagation invariant Mathieu beam. To measure the 3D intensity distribution, an optional mirror is placed in the focal plane of the microscope objective. By this means the reflected light is imaged onto a CMOS camera. Transversal intensity distributions of various Mathieu beams are measured by this method and depicted in Fig. 1 (lower rows) for direct comparison with the numerical calculations (upper rows in the figure). It is not expected and not observed that polarization has a significant influence on the focus shape with the used NA; the back aperture of the microscope objective was not completely filled and we estimate an

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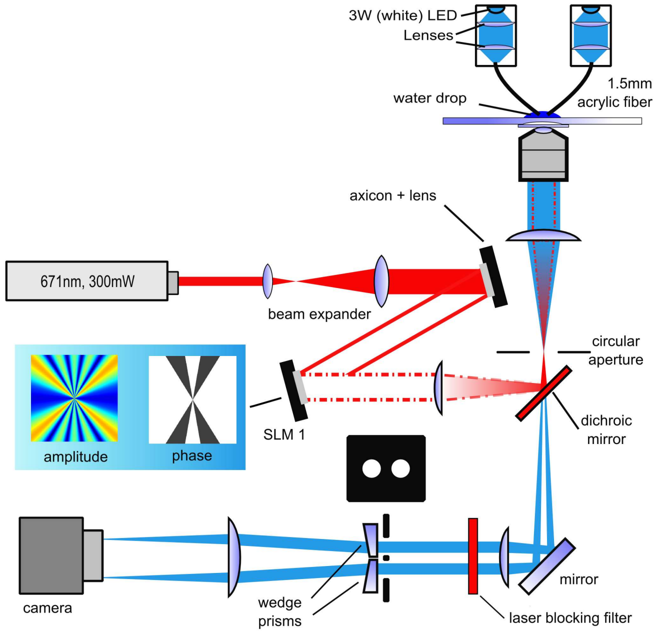

Fig. 3. Generation of optical light moulds: Light field tailoring setup for propagation invariant beams in stereoscopic tweezers.

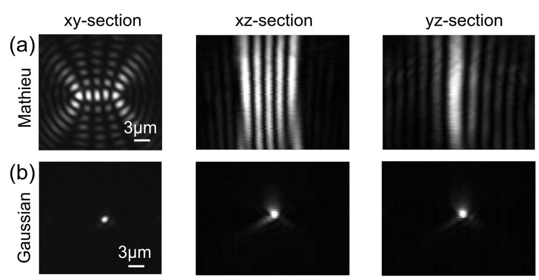

Fig. 4. Comparison between Mathieu beam (even, $m = 4$) and Gaussian beam: xy- (left), xz- (middle) and yz-sections (right). Distance in z-direction $\approx 11\ \mu$m; step size $\Delta z \approx 110$ nm.

effective NA of 80% of the objective's nominal NA value.

By scanning the z-position of the microscope objective, we are able to see the transversal intensity distribution in different planes. This gives xz- and yz-sections, showing the dimensions of the propagation invariant Mathieu beam. In Fig. 4 (a), these sections are shown for a fourth order ($m = 4$) even Mathieu beam. The propagation invariance could be observed here over a distance of about 11 μm in propagation direction (z-direction). Similar values were found for different Mathieu beams as the length of propagation invariance mainly depends on experimental parameters that were kept constant, such as the used NA and the thickness of the ring illuminating SLM 1. To further illustrate the propagation invariance of Mathieu beams, we directly compared them with a Gaussian beam (s. Fig. 4 (b)). The difference in their divergence can be clearly seen.

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4. Moulding of microparticles with Mathieu beams

To get an impression of the functionality of the available light mould, we measured the movement of a 2 μm silica probe within the 3D light field of a fourth order ($m = 4$) Mathieu beam. The probe is free to move along the direction of propagation as the intensity stays approximately constant, and is pushed upwards by the scattering force. When the probe reaches the top, the Mathieu beam is switched off and the particle is guided back to the bottom of the sample chamber. This procedure is repeated several times. The results of this measurement are stereoscopic images, from which the x,y,z-positions can be calculated. With the data of the online stereoscopic position detection, we can visualize the particle's path in 3D (s. Fig. 5 (a-b)). The identified potential consists of two minima, which fit to the two main intensity maxima of the related Mathieu beam. Parallel to the propagation direction, the potential is expanded along the optical axis (z-direction).

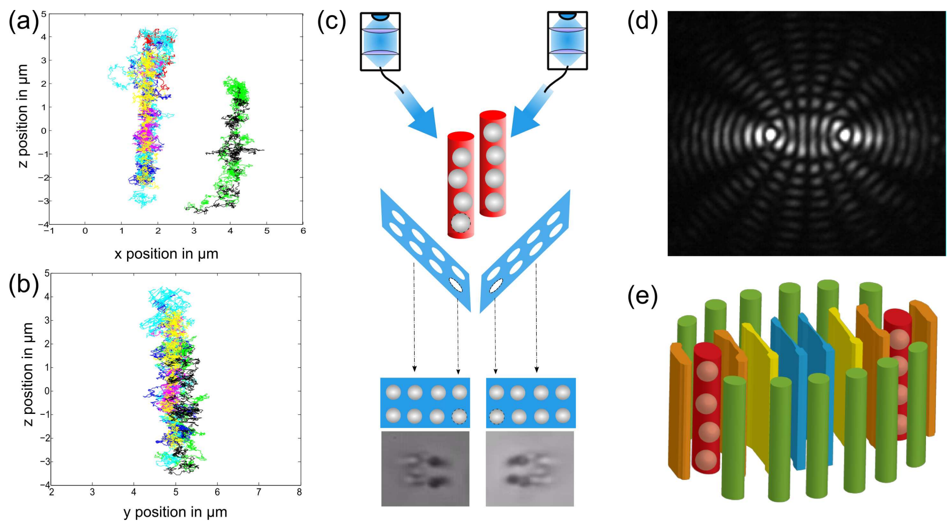

Fig. 5. Optical light mould characterization and material structuring. (a,b) Probe positions within the optical light mould of an $m = 4$ Mathieu beam. (c) Stereoscopic images of moulded particles (schematic + experiment) in an $m = 7$ Mathieu beam. (d) Transversal intensity distribution of the used Mathieu beam ($m = 7$). (e) Trapping configuration of 2 stacks of particles within the 3D intensity distribution.

To demonstrate the capability of the optical light mould, we trap 2 μm silica spheres in a seventh order ($m = 7$) Mathieu beam. Transversally, the spheres are arranged in the two main maxima of the Mathieu beam. There is no strong reason not to use an $m = 4$ Mathieu beam as before, but the particles are easier to observe if they are separated by a larger distance as in higher order mathieu beams. By tuning the laser power, it is possible to find a longitudinal equilibrium between scattering, gradient and gravitational forces. We are able to stack up to 6 2 μm spheres spheres (4 in this example) longitudinally in each of the two traps, because the self-healing beam is reconstructed after passing through each particle. With the stereoscopic microscope, we get a 3D view of the moulded 3D particle structure (Fig. 5 (c,e)). A most promising application for light moulds of this kind might be the arrangement of molecular hosts leading to hierarchical supramolecular organization [18].

The length of the shadow zone for 2 μm particles can be estimated at $l \leq 1.5\mu$m (from the center of the particle), if we assume we use 80% of the NA of the microscope objective. The distance between the centers of two stacked particles in the experiment can be estimated from

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the transversal displacement of the stereoscopic images. It is about 2 μm, which is the smallest possible displacement with respect to the particles' dimensions.

Basically the optical light moulds created by Mathieu beams should be suitable for most types of particles commonly used in optical tweezers experiments. Transparent particles are trapped on intensity maxima and absorbing or metallic particles avoid intensity maxima and e.g. can be confined between multiple maxima [19]. Obviously, the dimensions of particles must not exceed the characteristic feature size of the utilized Mathieu beam's intensity pattern. As gravitational forces are utilized to compensate for axial scattering forces, the relative mass density of particles needs to be in a reasonable range; very light particles might be pushed away and very heavy particles might sediment, if the dynamic range of laser light power is insufficient for adequate adaptation.

5. Three dimensional moulding of non-spherical objects

Various objects, such as bacteria in biology [20] and medicine or nano-containers such as zeolite L in material sciences [6] have non-spherical shapes. In single tweezers those particles with any distinctive axis always orientate parallel to the propagation direction of the light. Therefore, potential applications are limited. To trap elongated particles in a transverse plane, it has been shown, that one trap at each end of the particle would give a good trapping tool [21, 20]. In our second trapping configuration, we show trapping of elongated particles within

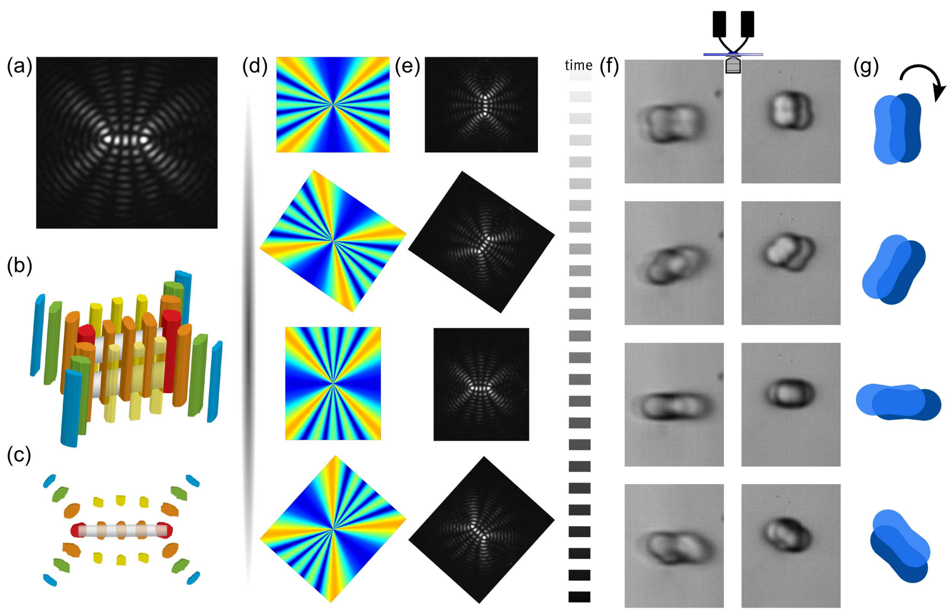

Fig. 6. 3D arrangements of elongated particles. (a) Mathieu beam $m = 4$ (experiment). (b) Trapping configuration of 2 stacked particles within 3D intensity distribution. (c) Particles orientation within transversal intensity distribution. (d) Rotating hologram. (e) Rotating Mathieu beam. (f) Time series of rotating 3D particle structure (stereoscopic)(Media 1).(g) Rotation of moulded particles.

a light mould, build by several intensity maxima of the transversal field distribution of a fourth order ($m = 4$) even Mathieu beam (Fig. 6(a)). Rod-like shaped silica particles (approx. 3 x 5 μm) are arranged with their long axis in the transversal plane of the Mathieu beam (Fig. 6 (c)). Here, the advantage of Mathieu beams over simpler self-healing beams like Bessel beams

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is particularly evident: their non-circular but elliptical symmetry makes them well suited for trapping and orienting elongated objects. The (even) fourth order beam is chosen as the feature size of the central maxima corresponds best to the particles' dimensions. Again, it is possible to stack multiple particles in z-direction because of the self-healing property of Mathieu beams. Every particle is arranged in the same transversal direction as the former (Fig. 6 (b)). This gives fascinating views for applications in hierarchical supramolecular organization experiments [6]. To achieve further complexity and diversity, we move from static configurations to dynamic configurations. In this approach we perform rotation of the structured material, which follows the rotation of the moulding light beam (Fig. 6 (d-g)). It is possible to move light fields and particle structures in real time within the x-y plane. In our experiment the movement is implemented by a rotation of the hologram. This demonstration gives promising applications in microfluidics, e.g. to build new 3D mixing tools for lab-on-a-chip devices.

6. Conclusion

In conclusion, optical light moulds are multifunctional tools for material structuring on microscopic scales, because they can be tailored in arbitrary shapes. By directly carrying their form to the material, they give a variety of future applications in areas as photonic band gab structures or metamaterials, microfluidics, hierarchical supramolecular organization, atom optics or 3D tissue engineering. We show the implementation of propagation invariant Mathieu beams as optical light moulds. In optical tweezers, we demonstrate three dimensional particle arrangements within the sample volume in different configurations: Stacking of 2 μm spheres in separated maxima of Mathieu beams; and by using the complex light mould of even Mathieu beams, arrange and rotate elongated particles in all three dimensions.

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